Critical Fluctuations in A Binary Polymer Mixture As Studied by Ultrasonic and Light Scattering Experiments*

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The critical mixture of polypropylene glycol (M = 1000 g/mol) and polyethylene glycol (M = 400 g/mol) was investigated by light scattering and ultrasonic experiments in the homogeneous one-phase region (explored temperature and frequency range of the ultrasonic experiment: $0.1~K \le T$ - $T_C \le 21.3~K$, $0.4~MHz \le f \le 30~MHz$, $T_C = 1.3~K$. Critical temperature). The composition of the critical mixture was determined by measuring the volume ratios of the separated phases in the inhomogeneous two-phase region (criterion of equal volumes).

The ultrasonic measurements are interpreted by a dynamic scaling theory for low molecular weight binary critical mixtures of Bhattacharjee and Ferrell [1]. The characteristic time scale of the dynamics of the critical concentration fluctuations is described by the frequency $\omega_c = 2D/\xi^2$ (D: mutual diffusion coefficient, ξ : correlation length), which can also be expressed by $\omega_C = \omega_O \, \epsilon^{zv} \, (\epsilon = (T-T_C)/T_C)$: reduced temperature, ω_O : critical amplitude, zv: critical exponent). The experimental values are $\omega_O = 22.2$ MHz from light scattering experiments and $\omega_O = 30$ MHz from ultrasonic data (within the frame of the Bhattacharjee-Ferrell theory). The low mutual diffusion coefficient of the mixture (D = $D_o \epsilon^{-v^*}$, v^* : critical exponent, D_O : critical amplitude, experimental value from dynamic light scattering $D_O = 4.0 \, 10^{-8} \, \text{cm}^2 \text{s}^{-1}$) allows to study the high frequency behavior of critical ultrasound attenuation in the range $10 < \Omega < 10^6 \, (\Omega = \omega/\omega_c)$: reduced frequency, ω : angular frequency of the measurement).

To get further insight into the physical mechanism of critical ultrasound behavior of a polymer blend, measurements of the thermodynamic coefficient dT_C/dP (pressure dependence of the critical temperature along the critical line) were performed [2]: $dT_C/dP \approx -5$ mK/bar. The value of $(\partial T/\partial P)_s$ (S: entropy) was calculated from thermodynamic parameters. The high frequency branch of the measured excess ultrasound attenuation was also compared to the theories of Kawasaki [3], Shiwa and Kawasaki [4,5] and Kroll and Ruhland [6]. The best agreement between data and theory was found for the predictions of dynamical scaling theory¹.

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